

# Multiphoton detachment of $H^-$ near the one-photon threshold: Exterior complex-scaling-generalized pseudospectral method for complex quasienergy resonances

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We perform a nonperturbative study of the multiphoton above-threshold detachment of  $H^-$  in the presence of 1.640- $\mu\text{m}$  and 1.908- $\mu\text{m}$  laser fields by means of the non-Hermitian Floquet formalism. The laser parameters used are related to the recent experiments [L. Præstegaard, T. Andersen, and P. Balling (unpublished)] on the two-photon detachment of  $H^-$  near the one-photon threshold. The total and partial (above-threshold) detachment rates as well as the electron angular distributions are calculated for the laser intensities from  $10^9 \text{ W/cm}^2$  to  $10^{12} \text{ W/cm}^2$ . It is found that at the weaker intensities (below  $10^{11} \text{ W/cm}^2$ ), the perturbation theory provides a reasonable description of the two-photon detachment process and the detached electrons are largely in the  $d$  state. For higher intensity, however, the process becomes highly nonperturbative in nature. To perform the calculations, we have introduced an exterior complex-scaling-generalized pseudospectral (ECS-GPS) technique for the discretization and solution of the non-Hermitian Floquet Hamiltonian. The ECS-GPS procedure is accurate, simple to implement, and computationally more efficient than the basis-set expansion-variational methods for resonance-state calculations. It also provides a simpler procedure than the uniform complex-scaling method for the calculations of partial rates and electron angular distributions.

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## I. INTRODUCTION

The study of multiphoton and above-threshold detachment processes of the  $H^-$  ion, a unique and important three-body atomic system, has attracted much interest both experimentally and theoretically in the past several years. The short-range interaction between the outer electron and the core supports only one bound state. Further, under the experimental conditions [1–5] for which the laser frequencies are either smaller than or comparable to the binding energy of the  $H^-$  ion, doubly excited states lie far above the detachment threshold and can be safely ignored. This simplifying feature renders the above-threshold multiphoton detachment of  $H^-$  a unique and fundamental process to study. The most recent experiments of  $H^-$  include the nonresonant [4,5] and resonant [6] two-photon above-threshold detachment observations. For the nonresonant cases [4,5], both the total detachment rates and the electron angular distributions have been measured.

Previous theoretical investigations of the processes of multiphoton detachment of  $H^-$  include the perturbation theory (see a summary in the paper by Geltman [7]), the Keldysh-Faisal-Reiss model [8], momentum-time-dependent calculations [9], inhomogeneous differential equation approach [10], two-electron perturbative or nonperturbative approaches [11–15], the nonperturbative time-independent non-Hermitian Floquet formalism [16–18], and the  $R$ -matrix Floquet method [19], to mention only a few. A theoretical study of the electron angular distribution and partial widths

for multiphoton above-threshold detachment of  $H^-$  (by 10.6- $\mu\text{m}$  laser field) was reported in one of our previous Floquet works [17].

The  $H^-$  ion will be described by an accurate one-electron model recently constructed [10] to reproduce both the exact experimental binding energy [20] and the low-energy  $e\text{-H}(1s)$  elastic scattering phase shifts. The one-photon detachment cross sections based on this model potential are in excellent agreement with earlier accurate two-electron calculations [21,22]. Using this model potential, Wang *et al.* [16] have performed detailed Floquet studies of the frequency- and intensity-dependent multiphoton detachment of  $H^-$ . The intensity-averaged multiphoton detachment rates and the threshold behavior so obtained are in good agreement with the previous Los Alamos experimental data [3], as well as the recent two-electron  $R$ -matrix Floquet calculation [23]. Finally, our recent Floquet study [18] of the electron angular distribution associated with the above-threshold multiphoton detachment of  $H^-$  by a 1064-nm laser field, again using this model potential, is in good harmony with the recent experimental work at Los Alamos [4]. In these recent Floquet studies [16–18], the (uniform) complex-scaling-generalized pseudospectral (CSGPS) method [16,24] is used for the discretization and solution of the non-Hermitian Floquet Hamiltonian. The CSGPS method is found to be both accurate and computationally efficient and is applicable to both low-lying and highly excited atomic and molecular resonance states.

The motivations of this paper are twofold. First, our present work on  $H^-$  is motivated by the most recent ongoing experiment in Denmark [5], which measures the electron angular distributions in the two-photon detachment of  $H^-$  near the one-photon threshold. Some preliminary results in this study were used by an experimental group [5] for the calibration of the experimental data. Second, we introduce an

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exterior complex-scaling–generalized pseudospectral (ECS-GPS) technique for accurate and efficient treatment of resonance states and apply the procedure to the study of complex quasienergy resonances associated with multiphoton above-threshold detachment of  $H^-$ . As will be shown later, the ECS-GPS technique has the advantage over the uniform CSGPS method in providing a simplified procedure for the calculation of partial rates and angular distributions.

The exterior complex-scaling transformation was proposed by Simon [25] for the treatment of molecular resonances in the Born-Oppenheimer approximation. It has been subsequently extended to the study of atomic and molecular resonances, particularly for potentials which behave nonanalytically (or defined only numerically or piecewise analytically) in the interior region of the coordinates. For such potentials, although the uniform complex scaling is still possible by means of certain transformation techniques [26,27], the exterior complex scaling provides a direct and alternative procedure. The principal idea of ECS is to perform the analytical continuation (complex scaling) of the coordinates beyond some distance  $R_b$  only. Thus for the one-particle system, the contour  $R(r)$  in the complex plane of the coordinate can be defined as follows:

$$R(r) = \begin{cases} r, & 0 \leq r \leq R_b \\ R_b + (r - R_b)\exp(i\alpha), & r > R_b. \end{cases} \quad (1)$$

Here  $r$  is assumed to be real valued while  $R(r)$  becomes complex valued beyond the radius  $R_b$ . For many-body systems, the same transformation is performed for each interparticle coordinate. A number of applications of the exterior complex-scaling procedure have been developed in the time-independent calculations of atomic and molecular resonances [28–32], cross sections in electron-atom collisions [33], as well as in time-dependent calculations [34]. Various numerical techniques were used to solve the second-order differential equation along the contour defined by Eq. (1): propagation and matching methods [28,31,32], global basis-set expansions [29], and finite-element basis-set expansions [30,34], etc. The function  $R(r)$  is not analytical at the point  $R_b$ , so some care should be taken when solving the equation along the contour (1). This issue is the most important for the basis-set variational calculations. The boundary conditions at the point  $R_b$  can be inserted in the Hamiltonian, leading to the appearance of an additional zero-range potential [29,35]. One can avoid the problem by introducing a discontinuous wave function transformation and a basis set of discontinuous functions [30,35] or using integration by parts in the kinetic energy matrix elements [36]. Certainly, the singular potential does not appear if the transition between the interior (unscaled) and exterior (complex scaled) regions of the coordinate is performed with an analytical function  $R(r)$  [32,37]. However, nontrivial mapping functions also complicate the problem, producing additional terms in the Hamiltonian.

In the present paper, we introduce an implementation of the exterior scaling method by means of the generalized pseudospectral technique [16,24,38], providing a simple yet highly accurate and efficient procedure. The *uniform* complex scaling within this GPS method was successfully applied for atomic and molecular resonance calculations (see,

e.g., [16–18,39,40]). According to this CSGPS approach, the complex-rotated coordinate is discretized on a set of collocation grid points, the potential matrix elements being diagonal and equal to the values of the potential at the grid points. The kinetic energy matrix elements have simple explicit analytical expressions. As discussed elsewhere [16,24], this uniform CGSPS procedure is found to be highly accurate and computationally more efficient than the traditional basis-set expansion method. For the *exterior* scaling, the whole range of the coordinate is split into two domains, the pseudospectral discretization being performed separately in each domain. The complex scaling is applied in the exterior domain only. The boundary conditions at the boundary point  $R_b$  can be incorporated in the discretized Hamiltonian, modifying the matrix elements. The new matrix elements also have simple explicit expressions, and the calculation of the Hamiltonian matrix in the generalized pseudospectral method with the exterior complex scaling is as simple as with the uniform complex scaling.

The paper is organized as follows. In Sec. II we describe the generalized pseudospectral technique with exterior complex scaling. In Sec. III we apply the ECS-GPS procedure to the study of multiphoton above-threshold detachment of  $H^-$  in connection with the recent experiment [5].

## II. EXTERIOR COMPLEX-SCALING–GENERALIZED PSEUDOSPECTRAL METHOD FOR MULTIPHOTON QUASIENERGY RESONANCES

In this section, we describe an approach, the exterior complex-scaling–generalized pseudospectral method, for accurate and efficient treatment of atomic and molecular resonances, including multiphoton quasienergy resonances (within the non-Hermitian Floquet Hamiltonian formalism [41,42]). The CSGPS method [16,24] is a natural extension of the complex-scaling generalized Fourier-grid Hamiltonian (CSGFGH) methods [43]. The CSGFGH methods employ Fourier series and require that mesh points be equally spaced. On the other hand, the CSGPS methods employ orthogonal polynomials (such as Legendre or Chebyshev polynomials) and allow for *nonuniform* grid spacing. It has been shown that the CSGFGH methods work well for potentials without singularity, such as the Morse potential for chemical bonds etc. [43,44]. However, for problems involving singularity and/or long-range potentials (such as the Coulomb potential), the CSGPS method with appropriate coordinate mapping [16,24] is the more natural and effective approach. In the following, we first review the essence of the GPS method and the uniform complex-scaling GPS method for bound- and resonance-state problems [16,24]. This is followed by a presentation of the ECS-GPS procedure.

### A. The generalized pseudospectral method with coordinate mapping for bound-state eigenvalue problems involving Coulomb potential

The central part of the pseudospectral method is to approximate the *exact* function  $f(x)$  defined on the interval  $[-1,1]$  by  $N$ th-order polynomial  $f_N(x)$ ,

$$f(x) \cong f_N(x) = \sum_{j=0}^N f(x_j) g_j(x), \quad (2)$$

and requires the approximation to be *exact* at the *collocation points*  $x_j$ :

$$f_N(x_j) = f(x_j). \quad (3)$$

In the case of the Legendre pseudospectral method which we shall employ in this article,  $x_0 = -1$ ,  $x_N = 1$ , and  $x_j$  (for  $j = 1, \dots, N-1$ ) are the collocation points determined by the roots of the first derivative of the Legendre polynomial  $P_N(x)$  with respect to  $x$ , namely,

$$P'_N(x_j) = 0. \quad (4)$$

In Eq. (2),  $g_j(x)$  are the cardinal functions defined by

$$g_j(x) = -\frac{1}{N(N+1)P'_N(x_j)} \frac{(1-x^2)P'_N(x)}{x-x_j} \quad (5)$$

and satisfy the unique property

$$g_j(x_{j'}) = \delta_{j'j}. \quad (6)$$

Consider the eigenvalue problem for the radial Schrödinger equation defined on the semi-infinite axis  $[0, \infty]$  with the Dirichlet boundary conditions:

$$\hat{H}(r)\psi(r) = E\psi(r), \quad \psi(0) = \psi(\infty) = 0, \quad (7)$$

where

$$\hat{H}(r) = -\frac{1}{2} \frac{d^2}{dr^2} + V(r). \quad (8)$$

For atomic structure and dynamics calculations involving the Coulomb potential, one typical problem with the grid methods is the Coulomb singularity at  $r=0$  and the long-range nature of the interaction. For *equal-spacing* grid methods, one generally truncates the semi-infinite domain into a finite domain  $[r_{\min}, r_{\max}]$ . For this purpose,  $r_{\min}$  must be chosen to be sufficiently small and  $r_{\max}$  sufficiently large. Furthermore, the grid spacing must also be chosen to be small enough to ensure that the short-range part of the Coulomb interaction is properly represented. This results in a need for a very large number of grid points, in addition to possible truncation errors. To overcome this problem, one can first map the semi-infinite domain  $r \in [0, \infty]$  into the finite domain  $x \in [-1, 1]$  using the mapping transformation

$$r = r(x), \quad (9)$$

and then use the Legendre or Chebyshev pseudospectral discretization techniques. In the previous works [16–18,24] for uniform complex scaling, we have used the following algebraic mapping:

$$r = r(x) = R_m \frac{1+x}{1-x}, \quad (10)$$

where  $R_m$  is a mapping parameter. Generally the introduction of nonlinear mapping can lead to either an asymmetric or a generalized eigenvalue problem. Such undesirable features can be avoided by the following symmetrization procedure [16,24]. Thus by introducing

$$\psi(r(x)) = \sqrt{r'(x)} f(x), \quad (11)$$

we obtain the following transformed Hamiltonian, leading to a *symmetric* eigenvalue problem (in atomic units):

$$\hat{H}(x) = -\frac{1}{2} \frac{1}{r'(x)} \frac{d^2}{dx^2} \frac{1}{r'(x)} + V(r(x)) + V_m(x), \quad (12)$$

where

$$V_m(x) = \frac{3(r'')^2 - 2r'''r'}{8(r')^4}. \quad (13)$$

Note that for the special mapping, Eq. (10),  $V_m(x) = 0$ . Discretizing the Hamiltonian operator, Eq. (12), by the pseudospectral method, leads to the following set of coupled linear equations:

$$\sum_{j=0}^N \left[ -\frac{1}{2} D_{j'j}^{(2)} + \delta_{j'j} V(r(x_j)) + \delta_{j'j} V_m(r(x_j)) \right] A_j = E A_{j'}, \quad j' = 1, \dots, N-1. \quad (14)$$

Here the coefficients  $A_j$  are related to the wave function values at the collocation points as

$$A_j = r'(x_j) f(x_j) [P_N(x_j)]^{-1} = [r'(x_j)]^{1/2} \psi(r(x_j)) [P_N(x_j)]^{-1}, \quad (15)$$

and the matrix  $D_{j'j}^{(2)}$ , representing the second derivative with respect to  $r$ , is given by

$$D_{j'j}^{(2)} = [r'(x_{j'})]^{-1} d_{j'j}^{(2)} [r'(x_j)]^{-1}, \quad (16)$$

where  $d_{j'j}^{(2)}$  is the second derivative of the cardinal function  $g_j(x)$  with respect to  $x$  (see the Appendix). The pseudospectral approximation for the first derivative of the wave function  $\psi(r)$  with respect to  $r$ , calculated at the points  $r(x_{j'})$ , can be expressed through the coefficients  $A_j$ :

$$\left. \frac{d\psi(r)}{dr} \right|_{r(x_{j'})} = P_N(x_{j'}) [r'(x_{j'})]^{-1/2} \sum_{j=0}^N D_{j'j}^{(1)} A_j, \quad j' = 0, \dots, N \quad (17)$$

with the matrix  $D_{j'j}^{(1)}$  given by

$$D_{j'j}^{(1)} = [r'(x_{j'})]^{-1/2} d_{j'j}^{(1)} [r'(x_j)]^{-1/2}, \quad (18)$$

where  $d_{j'j}^{(1)}$  is the first derivative of the cardinal function with respect to  $x$  (see the Appendix).

## B. Resonance-state complex eigenvalue problems: uniform and exterior complex scaling with generalized pseudospectral discretization

The GPS method described above can be extended to the resonance-state problems by means of the uniform complex-scaling method [16,24] or by the exterior complex-scaling method as described below. For the uniform complex scaling

[45],  $r \rightarrow r \exp(i\alpha)$ , we have previously used the following mapping transformation [16,24]:

$$r = R_m \frac{1+x}{1-x} \exp(i\alpha). \quad (19)$$

Here both parameters  $R_m$  and  $\alpha$  are real;  $R_m$  is the mapping parameter which determines the density of the grid points while  $\alpha$  is the complex rotation angle. Under this transformation, the semiaxis  $r \in [0, \infty]$  is rotated in the complex plane by the angle  $\alpha$  and then mapped to the interval  $x \in [-1, 1]$ . Note that for the transformation (19) the additional potential  $V_m(x)$  vanishes, so the Hamiltonian matrix in Eq. (14) takes the following simple form:

$$H_{j'j} = -\frac{1}{2} D_{j'j}^{(2)} + \delta_{j'j} V(r(x_j)), \quad j', j = 1, \dots, N-1 \quad (20)$$

with the Dirichlet boundary conditions taken into account.

Unlike the uniform scaling, the exterior complex scaling assumes that only the exterior part of the radial coordinate semiaxis is complex rotated. That means the total semiaxis  $[0, \infty]$  is divided in two domains,  $[0, R_b]$  and  $[R_b, \infty]$ , with the differential equation (12) to be solved separately in each domain. For the exterior domain, one can use the mapping transformation  $r_{\text{ex}}(x)$ , slightly different from Eq. (19):

$$r_{\text{ex}} = R_b + R_m \frac{1+x}{1-x} \exp(i\alpha), \quad (21)$$

while in the interior domain the linear map  $r_{\text{in}}(x)$ ,

$$r_{\text{in}} = \frac{1}{2} R_b (1+x), \quad (22)$$

serves the purpose. The boundary point  $R_b$ , as well as  $R_m$  and  $\alpha$  are the parameters of the transformations. Both the maps (21) and (22) do not generate the additional potential  $V_m(x)$ , and the sets of linear equations for the coefficients  $A_j$  in the interior and exterior domains read

$$\sum_{j=0}^{N_{\text{in}}} H_{j'j}^{\text{in}} A_j^{\text{in}} = E A_{j'}^{\text{in}}, \quad j' = 1, \dots, N_{\text{in}} - 1 \quad (23)$$

$$\sum_{j=0}^{N_{\text{ex}}} H_{j'j}^{\text{ex}} A_j^{\text{ex}} = E A_{j'}^{\text{ex}}, \quad j' = 1, \dots, N_{\text{ex}} - 1 \quad (24)$$

$N_{\text{in}}$  and  $N_{\text{ex}}$  being the numbers of collocation points in the interior and exterior domains, respectively. The Hamiltonian matrices  $H_{j'j}^{\text{in}}$  and  $H_{j'j}^{\text{ex}}$  in the interior and exterior domains have the form (20)

$$H_{j'j}^{\text{in}} = -\frac{1}{2} D_{j'j}^{(2),\text{in}} + \delta_{j'j} V(r_{\text{in}}(x_j^{\text{in}})), \quad j', j = 1, \dots, N_{\text{in}} - 1 \quad (25)$$

$$H_{j'j}^{\text{ex}} = -\frac{1}{2} D_{j'j}^{(2),\text{ex}} + \delta_{j'j} V(r_{\text{ex}}(x_j^{\text{ex}})), \quad j', j = 1, \dots, N_{\text{ex}} - 1. \quad (26)$$

Here  $x_j^{\text{in}}$  and  $x_j^{\text{ex}}$  are the collocation points for the interior and exterior domains, respectively, and the second derivative matrices  $D_{j'j}^{(2),\text{in}}$  and  $D_{j'j}^{(2),\text{ex}}$  are defined according to Eq. (16):

$$D_{j'j}^{(2),\text{in}} = [r'_{\text{in}}(x_j^{\text{in}})]^{-1} d_{j'j}^{(2)} [r'_{\text{in}}(x_j)]^{-1}, \quad (27)$$

$$D_{j'j}^{(2),\text{ex}} = [r'_{\text{ex}}(x_j^{\text{ex}})]^{-1} d_{j'j}^{(2)} [r'_{\text{ex}}(x_j)]^{-1}. \quad (28)$$

The Dirichlet boundary conditions at  $r=0$  and  $r=\infty$  imply that

$$A_0^{\text{in}} = A_{N_{\text{ex}}}^{\text{ex}} = 0. \quad (29)$$

However, there is no Dirichlet condition at the point  $r=R_b$  which corresponds to  $A_{N_{\text{in}}}^{\text{in}}$  and  $A_0^{\text{ex}}$ . One has to impose the continuity of the wave function and its first derivative instead. The wave function  $\psi(R_b)$  can be calculated with the help of Eq. (15). Applying Eq. (15) in the interior and exterior domains and equating the results, one obtains

$$[r'_{\text{in}}(1)]^{-1/2} P_{N_{\text{in}}}(1) A_{N_{\text{in}}}^{\text{in}} = [r'_{\text{ex}}(-1)]^{-1/2} P_{N_{\text{ex}}}(-1) A_0^{\text{ex}}. \quad (30)$$

Similarly, the calculation of the first derivative according to Eq. (17) leads to the following relation:

$$\begin{aligned} [r'_{\text{in}}(1)]^{-1/2} P_{N_{\text{in}}}(1) \sum_{j=1}^{N_{\text{in}}} D_{N_{\text{in}},j}^{(1),\text{in}} A_j^{\text{in}} \\ = [r'_{\text{ex}}(-1)]^{-1/2} P_{N_{\text{ex}}}(-1) \sum_{j=0}^{N_{\text{ex}}-1} D_{0,j}^{(1),\text{ex}} A_j^{\text{ex}}. \end{aligned} \quad (31)$$

With the help of Eqs. (30) and (31) one can express the coefficients  $A_{N_{\text{in}}}^{\text{in}}$  and  $A_0^{\text{ex}}$ , corresponding to the boundary point  $R_b$ , through the other coefficients  $A_j^{\text{in}}$  and  $A_j^{\text{ex}}$ :

$$A_{N_{\text{in}}}^{\text{in}} = -\frac{1}{\nu} \left[ \sum_{j=1}^{N_{\text{in}}-1} D_{N_{\text{in}},j}^{(1),\text{in}} A_j^{\text{in}} + (-1)^{N_{\text{ex}}-1} \mu \sum_{j=1}^{N_{\text{ex}}-1} D_{0,j}^{(1),\text{ex}} A_j^{\text{ex}} \right], \quad (32)$$

$$A_0^{\text{ex}} = \frac{1}{\nu} \left[ \sum_{j=1}^{N_{\text{ex}}-1} D_{0,j}^{(1),\text{ex}} A_j^{\text{ex}} + (-1)^{N_{\text{ex}}-1} \frac{1}{\mu} \sum_{j=1}^{N_{\text{in}}-1} D_{N_{\text{in}},j}^{(1),\text{in}} A_j^{\text{in}} \right], \quad (33)$$

where the constants  $\mu$  and  $\nu$  are defined as follows:

$$\mu = \left[ \frac{r'_{\text{in}}(1)}{r'_{\text{ex}}(-1)} \right]^{1/2}, \quad \nu = \frac{1}{4} \left[ \frac{N_{\text{in}}(N_{\text{in}}+1)}{r'_{\text{in}}(1)} + \frac{N_{\text{ex}}(N_{\text{ex}}+1)}{r'_{\text{ex}}(-1)} \right]. \quad (34)$$

Substituting the expressions (32) and (33) into the sets of equations (23) and (24), one eliminates the coefficients  $A_{N_{\text{in}}}^{\text{in}}$  and  $A_0^{\text{ex}}$  and obtains a closed matrix eigenvalue problem for the remaining coefficients:

$$\begin{aligned}
& \sum_{j=1}^{N_{\text{in}}-1} \left[ H_{j'j}^{\text{in}} - \frac{1}{\nu} H_{j',N_{\text{in}}}^{\text{in}} D_{N_{\text{in}},j}^{(1),\text{in}} \right] A_j^{\text{in}} \\
& + (-1)^{N_{\text{ex}}} \frac{\mu}{\nu} H_{j',N_{\text{in}}}^{\text{in}} \sum_{j=1}^{N_{\text{ex}}-1} D_{0,j}^{(1),\text{ex}} A_j^{\text{ex}} = E A_{j'}^{\text{in}}, \\
& j' = 1, \dots, N_{\text{in}} - 1 \\
& \sum_{j=1}^{N_{\text{ex}}-1} \left[ H_{j'j}^{\text{ex}} + \frac{1}{\nu} H_{j',0}^{\text{ex}} D_{0,j}^{(1),\text{ex}} \right] A_j^{\text{ex}} \\
& - (-1)^{N_{\text{ex}}} \frac{1}{\mu \nu} H_{j',0}^{\text{ex}} \sum_{j=1}^{N_{\text{in}}-1} D_{N_{\text{in}},j}^{(1),\text{in}} A_j^{\text{in}} = E A_{j'}^{\text{ex}}, \\
& j' = 1, \dots, N_{\text{ex}} - 1.
\end{aligned} \tag{35}$$

As one can see, taking into account the boundary conditions at the point  $R_b$  modifies the Hamiltonian matrices in the interior and exterior domains as well as adds coupling matrix elements between the two domains. The total matrix of the eigenvalue problem (35) has the dimensions  $(N_{\text{in}} + N_{\text{ex}} - 2) \times (N_{\text{in}} + N_{\text{ex}} - 2)$ . The diagonalization of this matrix yields the eigenvalues and the eigenvectors  $\{A_j^{\text{in}}\}$  and  $\{A_j^{\text{ex}}\}$  inside the interior and exterior domains. Then the coefficients  $A_{N_{\text{in}}}^{\text{in}}$  and  $A_0^{\text{ex}}$ , corresponding to the boundary point  $R_b$ , can be obtained with the help of Eqs. (32) and (33).

### C. Exterior complex-scaling—generalized pseudospectral method for complex quasienergy resonances associated with multiphoton ionization processes

In the presence of periodically time-dependent fields, the time-dependent Schrödinger equation can be transformed into an equivalent *time-independent* infinite-dimensional Floquet Hamiltonian  $\hat{H}^F(\mathbf{r})$  eigenvalue problem [41,42]:

$$\hat{H}^F(\mathbf{r}) \psi(\mathbf{r}) = \varepsilon \psi(\mathbf{r}), \tag{36}$$

where  $\varepsilon$  is the quasienergy. The Floquet Hamiltonian  $\hat{H}^F$  has no discrete spectrum. Writing the time-evolution operator as

$$\exp(-i\hat{H}^F t) = \frac{1}{2\pi i} \int_C dz \frac{\exp(-izt)}{z - \hat{H}^F} \tag{37}$$

gives the usual result that the time dependence is dominated by poles of the resolvent  $(z - \hat{H}^F)^{-1}$  near the real axis but on higher Riemann sheets. The complex energies of the poles are related to the positions and widths  $(E_R, -\Gamma/2)$  of the shifted and broadened complex quasienergy states. In the non-Hermitian Floquet formalism [41,42], the complex quasienergy states are determined directly from the eigenvalue analysis of the analytically continued Floquet Hamiltonian  $\hat{H}^F(\mathbf{r} \exp(i\alpha))$ , obtained by the uniform complex-scaling transformation  $\mathbf{r} \rightarrow \mathbf{r} \exp(i\alpha)$  [45]. The non-Hermitian Floquet matrix can be solved by means of the  $L^2$  basis-set expansion—variational method [41,42] or by the CSGFGH [43,24] or CSGPS method [16–18]. In the present work, we extend the ECS–GPS procedure described in Sec. II for the

discretization and solution of the non-Hermitian Floquet Hamiltonian eigenvalue problem. The procedure is similar to the uniform CSGPS method [16–18], except the radial distance  $r$  is partitioned into the interior and exterior regions, Eq. (1). The optimal distance  $R_b$  to partition the two regions can be chosen in such a way that the dominant interaction potential is included in the interior region while the exterior region includes only the long-range part of the interaction. Further, as will be shown in the next section, the calculation of the partial rates and angular distributions can be basically accomplished within the interior region, resulting in a significant simplification of the overall procedure.

### III. CALCULATIONS OF PARTIAL RATES AND ELECTRON ANGULAR DISTRIBUTIONS FOR MULTIPHOTON DETACHMENT OF $\text{H}^-$ NEAR ONE-PHOTON THRESHOLD

In this section we apply the ECS-GPS method to the solution of the non-Hermitian Floquet Hamiltonian associated with multiphoton detachment of  $\text{H}^-$  by 1.640- $\mu\text{m}$  and 1.908- $\mu\text{m}$  laser fields corresponding to the recent experiments ongoing in Denmark [5]. The procedure for the calculation of electron energy and angular distributions within the Floquet formalism has been described elsewhere [18]. Here we outline the basic formulas for the description of multiphoton detachment of  $\text{H}^-$ .

We describe the unperturbed  $\text{H}^-$  negative ion within the accurate single-electron model potential approximation [10]. In the presence of linearly polarized monochromatic fields, the total electron wave function, according to the Floquet theorem, can be written as

$$\Psi(\mathbf{r}, t) = \exp(-i\varepsilon t) \psi(\mathbf{r}, t), \tag{38}$$

where  $\varepsilon$  is the quasienergy. The function  $\psi(\mathbf{r}, t)$  is periodic in time with the period  $T = 2\pi/\omega$  and can be expanded in the Fourier series:

$$\psi(\mathbf{r}, t) = \sum_{m=-\infty}^{\infty} \psi_m(\mathbf{r}) \exp(-im\omega t). \tag{39}$$

The Fourier components  $\psi_m(\mathbf{r})$  constitute an infinite-dimensional vector which solves the eigenvalue problem for the time-independent Floquet Hamiltonian  $\hat{H}^F$  [41,42]:

$$\sum_{m'} \hat{H}_{mm'}^F \psi_{m'}(\mathbf{r}) = \varepsilon \psi_m(\mathbf{r}). \tag{40}$$

The diagonal blocks  $\hat{H}_{mm}^F$  of the Floquet Hamiltonian are the energy-shifted unperturbed Hamiltonian operators for the electron moving in the  $\text{H}^-$  model potential:

$$\hat{H}_{mm}^F = -\frac{1}{2} \nabla^2 + \hat{W}(\mathbf{r}) - m\omega, \tag{41}$$

where  $\hat{W}(\mathbf{r})$  is the angular-momentum-dependent model potential [10]. The off-diagonal blocks of the Floquet Hamiltonian represent the interaction of the electron with the external laser field. The quasienergy eigenvalues emerging from the eigenvalue problem (40) are complex valued,

( $E_R, -\Gamma/2$ ). The real part of the complex quasienergy ( $E_R$ ) provides the ac Stark shifted energy level, while  $\Gamma$  gives rise to the total multiphoton ionization rate.

The expression for the electron angular distributions after absorption of  $n$  linearly polarized photons can be written as [18]

$$\frac{d\Gamma_n}{d\Omega} = (2\pi)^{-2} k_n |A_n|^2. \quad (42)$$

Here,

$$k_n = \sqrt{2[\text{Re } \varepsilon - (2\omega)^{-2} F^2 + n\omega]} \quad (43)$$

is the electron drift momentum, and the  $n$ -photon detachment amplitude  $A_n$  is defined as follows:

$$\begin{aligned} A_n = & (2\pi)^{-1} \int_{-\pi}^{\pi} d\tau \exp[in\tau - i(2\omega)^{-3} F^2 \sin(2\tau) \\ & + ik_n(\hat{\mathbf{r}} \cdot \mathbf{F}) \omega^{-2} \cos \tau] \int d^3\mathbf{r}' \exp[-ik_n(\hat{\mathbf{r}} \cdot \mathbf{r}') \\ & + i(\mathbf{r}' \cdot \mathbf{F}) \omega^{-1} \sin \tau] \hat{W}(\mathbf{r}') \psi(\mathbf{r}', \tau/\omega), \end{aligned} \quad (44)$$

$\mathbf{F}$  and  $\omega$  being the laser field strength and frequency, respectively. Equations (42) and (44) assume that the wave function  $\psi(\mathbf{r}, t)$  is properly normalized. The expression (44) is suitable for practical computations since the integration over the angles in the spatial integral can be performed analytically, and the integral over the  $\tau$  variable can be computed effectively using the fast Fourier transform routines. The quantity  $d\Gamma_n/d\Omega$  represents the number of electrons per unit time detached with absorption of  $n$  photons and emitted within the unit solid angle under direction of the unit vector  $\hat{\mathbf{r}}$ . Integration of the angular distributions (42) with respect to the angles specifying the direction  $\hat{\mathbf{r}}$  gives the partial rates  $\Gamma_n$ :

$$\Gamma_n = \int d\Omega \frac{d\Gamma_n}{d\Omega}. \quad (45)$$

The sum of all partial rates with  $n \geq n_{\min}$ , where  $n_{\min}$  is the minimum number of photons required for detachment, is equal to the total rate  $\Gamma$ :

$$\Gamma = \sum_{n=n_{\min}}^{\infty} \Gamma_n. \quad (46)$$

One can expand  $d\Gamma_n/d\Omega$  as a function of the angle  $\theta$  between the detection  $\hat{\mathbf{r}}$  and field  $\mathbf{F}$  directions on the basis of the Legendre polynomials. Due to parity restrictions, only even Legendre polynomials are present in the expansion:

$$\frac{d\Gamma_n}{d\Omega} = \frac{\Gamma_n}{4\pi} \left( 1 + \sum_{l=1}^{\infty} \beta_{2l} P_{2l}(\cos \theta) \right). \quad (47)$$

The coefficients  $\beta_{2l}$  are the anisotropy parameters since they determine the deviation of the real angular distribution from the isotropic one. When analyzing the behavior of the coefficients  $\beta_{2l}$  for weak and medium-strong external fields, a

comparison with the results of the lowest-order perturbation theory (LOPT) is valuable. For the one-photon detachment the prediction of the perturbation theory is  $\beta_2=2, \beta_{2l}=0$  ( $l>1$ ). The situation is more complicated if the number of absorbed photons  $n=2$ . According to LOPT, the emitted electrons in this case may possess the angular momentum 0 or 2. For the emitted electron in the pure  $d$  state, one has  $\beta_2=10/7, \beta_4=18/7, \beta_{2l}=0$  ( $l>2$ ) whereas for the pure  $s$  state the distribution is isotropic, i.e., all  $\beta_{2l}=0$ . In reality, however,  $s$  and  $d$  waves are mixed in the wave function of the emitted electron, so the coefficients  $\beta_{2l}$  cannot be calculated with pure angular algebra even for the lowest intensities. According to LOPT, the detachment amplitude should behave as

$$\delta \sqrt{\frac{1}{2}} P_0(\cos \theta) + \sqrt{\frac{5}{2}} P_2(\cos \theta), \quad (48)$$

i.e., it contains contributions from  $s$  and  $d$  partial waves. The factors  $\sqrt{1/2}$  and  $\sqrt{5/2}$  are added as normalization coefficients for the Legendre polynomials. The mixing coefficient  $\delta$  can be calculated within LOPT: in general it depends not only on the angular algebra, but also on the radial wave functions. Squaring the absolute value of the amplitude written above and expanding it over the even-order Legendre polynomials, one obtains for the coefficients  $\beta_{2l}$

$$\begin{aligned} \beta_2 &= \frac{10 + 14 \text{Re } \delta \sqrt{5}}{7(1 + |\delta|^2)}, \\ \beta_4 &= \frac{18}{7(1 + |\delta|^2)}, \end{aligned} \quad (49)$$

other coefficients being zero within LOPT. Given the mixing parameter  $\delta$ , one can calculate the anisotropy parameters  $\beta_2$  and  $\beta_4$ . For example, if one sets  $\delta=0$  (pure  $d$  wave in the final state), the results 10/7 and 18/7 mentioned above are obtained. On the other hand, if we take the  $\beta_2$  and  $\beta_4$  coefficients from our calculations, we can find the complex mixing parameter  $\delta$ :

$$\begin{aligned} \text{Re } \delta &= \frac{9\beta_2/\beta_4 - 5}{7\sqrt{5}}, \\ \text{Im } \delta &= \sqrt{\frac{18}{7\beta_4} - 1 - (\text{Re } \delta)^2}. \end{aligned} \quad (50)$$

The coefficient  $\delta$  calculated in this way is intensity dependent. In the limit of the weak external field this result should converge to the intensity independent value which can be determined within LOPT.

We have performed the calculations of the total and partial (above threshold) multiphoton detachment rates as well as the angular distributions of the emitted electrons. Equation (40) is solved with the help of the ECS-GPS technique described in Sec. II. The Fourier components  $\psi_m(\mathbf{r})$  of the wave function are further expanded in terms of the angular momentum eigenfunctions (the Legendre polynomials). Only the diagonal blocks of the Floquet Hamiltonian (with respect to the Fourier index  $m$  and angular momentum  $l$ ) are modi-

TABLE I. Partial and total rates for the detachment of  $H^-$  by  $1.640\text{-}\mu\text{m}$  radiation. The numbers in brackets indicate the powers of 10.

Laser intensity (W/cm <sup>2</sup> )	Partial rates (a.u.)					Total rates (a.u.)
	Number of photons absorbed					
	1	2	3	4	5	
1.0[9]	3.316[−9]	3.010[−9]	7.155[−13]	1.553[−16]		6.327[−9]
1.0[10]		2.978[−7]	7.099[−10]	2.087[−12]	9.706[−15]	2.985[−7]
1.0[11]		2.717[−5]	6.753[−7]	1.336[−8]	2.643[−10]	2.786[−5]
2.0[11]		9.938[−5]	5.134[−6]	2.109[−7]	8.655[−9]	1.047[−4]
4.0[11]		3.353[−4]	3.695[−5]	3.185[−6]	2.708[−7]	3.757[−4]
8.0[11]		9.482[−4]	2.316[−4]	4.202[−5]	7.339[−6]	1.229[−3]
1.0[12]		1.231[−3]	3.926[−4]	9.064[−5]	1.981[−5]	1.734[−3]

fied by the exterior complex-scaling procedure according to Eq. (35). The off-diagonal blocks are not affected except for the different mapping functions used in the interior and exterior domains of the radial coordinate semiaxis. In practical computations, the results are converged for the following ranges of the parameters:  $R_b = 40\text{--}60$  a.u.,  $R_m = 80\text{--}100$  a.u.,  $\alpha = 0.4\text{--}0.6$  radians. To achieve convergence, we include the Fourier components  $-12$  to  $12$  and angular momenta  $0$  to  $9$  for the highest intensity considered. Up to  $100$  radial grid points were used in each interior and exterior domain. The selected eigenvalue and eigenvector of the non-Hermitian Floquet Hamiltonian matrix can be obtained efficiently by means of the implicitly restarted Arnoldi algorithm [46] with spectral transformation. We found that the interior domain appears large enough to get the integral (44) fully converged within it. That means we do not need to perform the integration in the complex-rotated exterior domain using the *back rotation* procedure as in the uniform complex-scaling case [17]. This simplifies considerably the calculation of partial rates and angular distributions.

The calculations were performed for the laser field intensities in the range  $10^9\text{--}10^{12}\text{ W}/\text{cm}^2$  and the wavelengths  $1.640\text{ }\mu\text{m}$  and  $1.908\text{ }\mu\text{m}$ . The field parameters correspond to those used in the recent experiments on the electron angular distributions [5]. For the wavelength  $1.640\text{ }\mu\text{m}$ , the photon energy ( $\omega = 0.756\text{ eV}$ ) is very close

to the one-photon detachment threshold ( $0.754\text{ eV}$ ). The one-photon channel is open only for the weak intensity  $10^9\text{ W}/\text{cm}^2$ ; for the higher intensities it is closed due to ac Stark shift. In the  $1.908\text{-}\mu\text{m}$  ( $\omega = 0.650\text{ eV}$ ) field, a minimum of two photons is required for detachment for all the intensities used in the calculations. The results are presented in Tables I–III. (As an independent check of the reliability and accuracy of the ECS-GPS method, we have also performed the calculations using the uniform CSGPS technique. The results are essentially identical to those shown in Tables I–III.) Tables I and II contain the (above-threshold) partial and total detachment rates for the detachment by  $1.640\text{-}\mu\text{m}$  and  $1.908\text{-}\mu\text{m}$  radiation, respectively. One can see that the detachment rates for the same intensity are generally larger for the wavelength  $1.908\text{ }\mu\text{m}$ . The exception is made by the intensity  $10^9\text{ W}/\text{cm}^2$ , where the total detachment rate at the wavelength  $1.640\text{ }\mu\text{m}$  is larger than that at the wavelength  $1.908\text{ }\mu\text{m}$ . The difference is due to the one-photon contribution which is present for the  $1.640\text{-}\mu\text{m}$  field and not for the  $1.908\text{-}\mu\text{m}$  field. The above-threshold contribution is not very significant for the intensities up to  $10^{11}\text{ W}/\text{cm}^2$ . For example, for the intensity  $10^{11}\text{ W}/\text{cm}^2$ , the three-photon detachment rate constitutes only slightly more than 2% of the two-photon rate. The total detachment rates for the same wavelength and different intensities follow the power law of

TABLE II. Partial and total rates for the detachment of  $H^-$  by  $1.908\text{-}\mu\text{m}$  radiation. The numbers in brackets indicate the powers of 10.

Laser intensity (W/cm <sup>2</sup> )	Partial rates (a.u.)Total rates (a.u.)				Total rates (a.u.)
	Number of photons absorbed				
	2	3	4	5	
1.0[9]	4.846[−9]	2.211[−12]	8.278[−16]	3.054[−19]	4.848[−9]
1.0[10]	4.778[−7]	2.191[−9]	8.223[−12]	3.037[−14]	4.800[−7]
1.0[11]	4.165[−5]	1.996[−6]	7.650[−8]	2.844[−9]	4.373[−5]
2.0[11]	1.434[−4]	1.433[−5]	1.118[−6]	8.346[−8]	1.589[−4]
4.0[11]	4.231[−4]	9.070[−5]	1.453[−5]	2.162[−6]	5.304[−4]
8.0[11]	9.195[−4]	4.137[−4]	1.416[−4]	4.155[−5]	1.516[−3]
1.0[12]	1.126[−3]	6.353[−4]	3.008[−4]	1.138[−4]	2.176[−3]

TABLE III. Anisotropy parameters  $\beta_{2l}$  for the two- and three-photon detachment and the mixing parameters  $\delta$  for the two-photon detachment. The numbers in brackets indicate the powers of 10.

$l$	Number of absorbed photons			
	2	3	2	3
	1.640 $\mu\text{m}$		1.908 $\mu\text{m}$	
		$1 \times 10^9 \text{ W/cm}^2$		
1	1.153	2.381	6.278[−1]	2.359
2	2.505	2.709	2.298	2.407
3	−3.401[−4]	2.640	−4.119[−4]	2.313
4	2.291[−8]	−5.011[−4]	6.065[−8]	−6.347[−4]
	$\delta=0.1629 \exp(i1.914)$		$\delta=0.3449 \exp(i2.061)$	
		$1 \times 10^{10} \text{ W/cm}^2$		
1	1.134	2.369	6.137[−1]	2.362
2	2.499	2.687	2.291	2.404
3	−3.385[−3]	2.630	−4.076[−3]	2.301
4	1.781[−6]	−4.932[−3]	2.707[−6]	−6.305[−3]
	$\delta=0.1707 \exp(i1.921)$		$\delta=0.3496 \exp(i2.064)$	
		$1 \times 10^{11} \text{ W/cm}^2$		
1	1.068	2.398	4.623[−1]	2.375
2	2.454	2.650	2.226	2.375
3	−3.188[−2]	2.502	−3.680[−2]	2.198
4	1.599[−4]	−4.677[−2]	2.218[−4]	−5.937[−2]
	$\delta=0.2189 \exp(i1.893)$		$\delta=0.3937 \exp(i2.104)$	
		$2 \times 10^{11} \text{ W/cm}^2$		
1	9.855[−1]	2.415	2.681[−1]	2.372
2	2.410	2.608	2.152	2.337
3	−5.995[−2]	2.388	−6.549[−2]	2.109
4	5.815[−4]	−8.871[−2]	7.823[−4]	−1.122[−1]
	$\delta=0.2591 \exp(i1.902)$		$\delta=0.4415 \exp(i2.167)$	
		$4 \times 10^{11} \text{ W/cm}^2$		
1	8.024[−1]	2.427	−2.014[−1]	2.333
2	2.334	2.536	1.974	2.251
3	−1.066[−1]	2.214	−9.996[−2]	1.985
4	1.952[−3]	−1.624[−1]	2.098[−3]	−2.034[−1]
	$\delta=0.3191 \exp(i1.962)$		$\delta=0.5502 \exp(i2.328)$	
		$8 \times 10^{11} \text{ W/cm}^2$		
1	3.165[−1]	2.384	−1.272	2.160
2	2.186	2.389	1.304	2.077
3	−1.644[−1]	1.992	−7.867[−2]	1.909
4	5.105[−3]	−2.820[−1]	2.021[−3]	−3.493[−1]
	$\delta=0.4199 \exp(i2.168)$		$\delta=0.9858 \exp(i2.675)$	
		$1 \times 10^{12} \text{ W/cm}^2$		
1	5.106[−3]	2.341	−1.465	2.175
2	2.092	2.316	7.847[−1]	2.076
3	−1.750[−1]	1.924	−3.971[−2]	1.840
4	6.085[−3]	−3.322[−1]	8.638[−4]	−3.922[−1]
	$\delta=0.4786 \exp(i2.298)$		$\delta=1.509 \exp(i2.747)$	

LOPT quite closely (except for the  $10^9\text{-W/cm}^2, 1.640\text{-}\mu\text{m}$  case mentioned above). One can draw a conclusion that for the wavelength range under consideration the perturbation theory provides an adequate description of the detachment

process for the intensities up to  $10^{11} \text{ W/cm}^2$ . For the higher intensities, the contribution of the above-threshold channels to the total rate (i.e., deviation from the LOPT predictions) becomes very important. As can be expected from the gen-



eral theory, at the same intensity, the breakdown of the perturbation theory is more pronounced for the larger wavelength 1.908  $\mu\text{m}$ .

This conclusion is confirmed by the analysis of the angular distributions. Table III contains the anisotropy parameters  $\beta_{2l}$  for the two- and three-photon detachment as well as the mixing parameters  $\delta$  for the two-photon detachment. One can estimate how close the calculated nonperturbative results and those of LOPT are by taking a look at the magnitude of the coefficient  $\beta_6$  for the two-photon process and  $\beta_8$  for the three-photon process. These coefficients are supposed to vanish within LOPT. As expected, the smallest values of these coefficients correspond to the lowest intensity. With the increase of the laser intensity, their relative magnitude increases from approximately  $10^{-4}$  for  $10^9 \text{ W/cm}^2$  to  $10^{-1}$  for  $10^{12} \text{ W/cm}^2$ . As one can see, the angular distribution pattern is quite stable and the mixing parameter  $\delta$  is small for the intensities less than or equal to  $10^{11} \text{ W/cm}^2$ . For the laser fields with the wavelength 1.640  $\mu\text{m}$ , only 2.5% to 4.5% (depending on the field intensity) of the electrons emitted after absorption of two photons appear in the  $s$  state, all others being in the  $d$  state. For the wavelength 1.908  $\mu\text{m}$ ,  $s$  electrons constitute 10% to 13% of the total emission, and the rest is due to the  $d$ -electron contribution.

This picture changes rapidly as the laser intensity increases above  $10^{11} \text{ W/cm}^2$ . The set of the anisotropy parameters  $\beta_{2l}$  for the two-photon detachment at the intensities  $2 \times 10^{11} \text{ W/cm}^2$  to  $10^{12} \text{ W/cm}^2$  differs very much from that at the intensities  $10^9 \text{ W/cm}^2$  to  $10^{11} \text{ W/cm}^2$ . The mixing parameter  $\delta$  suggests that up to 20% of the electrons at the wavelength 1.640  $\mu\text{m}$ , and 70% of the electrons at the wavelength 1.908  $\mu\text{m}$ , are emitted in the  $s$  state for the laser field intensity  $10^{12} \text{ W/cm}^2$ . One must realize, however, that for such intense laser fields the parameter  $\delta$  does not have direct meaning since its definition is based on the LOPT expression (48) and can be interpreted only approximately. The accurate information is provided by the anisotropy parameters  $\beta_{2l}$ .

The recent experiment [5] provides the following values of the anisotropy parameters  $\beta_2$  and  $\beta_4$  for the two-photon detachment:

$$\beta_2 = 1.3 \pm 0.2, \quad \beta_4 = 2.4 \pm 0.2 \quad (1.640 \mu\text{m}), \quad (51)$$

$$\beta_2 = 0.7 \pm 0.2, \quad \beta_4 = 2.0_{-0.2}^{+0.4} \quad (1.908 \mu\text{m}). \quad (52)$$

As one can see from comparison with Table III, our nonperturbative results lie within the error range of the experimental values for intensities of the laser field less than or equal to  $10^{11} \text{ W/cm}^2$ , i.e., in that region where the deviation from the perturbation theory is not significant. The authors of Ref. [5] indicate that the peak intensity in their experiment can be as high as  $8 \times 10^{11} \text{ W/cm}^2$ . This intensity corresponds to the highly nonperturbative regime where the angular distributions are quite different from those for the intensities weaker than  $10^{11} \text{ W/cm}^2$  (see Table III). To explain this discrepancy, we can infer that under the experimental conditions [5] (the laser pulse duration is as long as 9 ns) the detachment process is likely to occur before the laser field pulse reaches the peak intensity and thus corresponds to the weaker intensity case.

In conclusion, we have presented an ECS-GPS general procedure for accurate and efficient determination of resonance-state properties with application to the study of complex quasienergy states associated with the multiphoton above-threshold detachment of  $\text{H}^-$  near the one-photon threshold. Both total and partial detachment rates are presented for laser wavelengths 1.640  $\mu\text{m}$  and 1.908  $\mu\text{m}$ , and for intensity in the range  $10^9 \text{ W/cm}^2$  to  $10^{12} \text{ W/cm}^2$ . It is found that for laser intensity less than  $10^{11} \text{ W/cm}^2$ , the two-photon detachment processes are dominated by the  $d$ -wave electrons, in accord with the recent experimental observations [5]. We are currently extending the ECS-GPS technique to the solution of the non-Hermitian Floquet Hamiltonian associated with the *time-dependent density functional theory* [47] for the study of multiphoton processes of *many-electron* systems in intense laser fields.

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### APPENDIX: PSEUDOSPECTRAL TECHNIQUE FOR THE SECOND-ORDER DIFFERENTIAL EQUATION EIGENVALUE PROBLEM

Consider an eigenvalue problem defined by a second-order differential equation

$$f''(x) + v(x)f(x) = \lambda f(x) \quad (A1)$$

on the interval  $[-1, 1]$ , with appropriate boundary conditions at  $x = -1$  and  $x = 1$  ( $\lambda$  is the eigenvalue). An approximate solution of the eigenvalue problem can be obtained with the help of the pseudospectral interpolation formula (2). Certainly, the polynomial  $f_N(x)$  cannot solve the differential equation (A1) for all  $x$  within the interval  $[-1, 1]$ . However, one can request that Eq. (A1) is satisfied at the collocation points  $x_j$  upon substitution of  $f_N(x)$  for  $f(x)$ . One then arrives at a set of linear algebraic equations for the quantities  $f(x_j)$ :

$$\sum_{j=0}^N f(x_j) [g_j''(x_{j'}) + v(x_{j'})g_j(x_{j'}) - \lambda g_j(x_{j'})] = 0,$$

$$j' = 1, \dots, N-1. \quad (A2)$$

Note that the set (A2) contains just  $N-1$  equations for  $N+1$  coefficients  $f(x_j)$ . It must be appended by two more equations representing the boundary conditions at  $x = -1$  and  $x = 1$ , defining the algebraic matrix eigenvalue problem.

The derivatives of the functions  $g_j(x)$  can be calculated as follows (see [16,24,38]):

$$g_j'(x_{j'}) = d_{j'j}^{(1)} \frac{P_N(x_{j'})}{P_N(x_j)}, \quad (A3a)$$

$$d_{j'j}^{(1)} = \frac{1}{x_{j'} - x_j} \quad (j' \neq j), \quad d_{jj}^{(1)} = 0 \quad (j \neq 0, j \neq N), \quad (A3b)$$

$$d_{00}^{(1)} = -\frac{N(N+1)}{4}, \quad d_{NN}^{(1)} = \frac{N(N+1)}{4}, \quad (A3c)$$

$$g_j''(x_{j'}) = d_{j'j}^{(2)} \frac{P_N(x_{j'})}{P_N(x_j)}, \quad (A4a)$$

$$d_{j'j}^{(2)} = -\frac{2}{(x_{j'} - x_j)^2} [j' \neq j, (j'j) \neq (0N), (j'j) \neq (N0)], \quad (A4b)$$

$$d_{0N}^{(2)} = d_{N0}^{(2)} = \frac{N(N+1)-2}{4}, \quad (A4c)$$

$$d_{jj}^{(2)} = -\frac{N(N+1)}{3(1-x_j^2)} \quad (j \neq 0, j \neq N), \quad (A4d)$$

$$d_{00}^{(2)} = d_{NN}^{(2)} = \frac{N(N+1)[N(N+1)-2]}{24}. \quad (A4e)$$

Let us introduce the coefficients  $A_j$ , related to  $f(x_j)$  as

$$f(x_j) = P_N(x_j)A_j. \quad (A5)$$

Taking into account the relations (6), (A4), and (A5), one can recast the set of equations (A2) in the following form:

$$\sum_{j=0}^N [d_{j'j}^{(2)} + \delta_{j'j} v(x_j)] A_j = \lambda A_{j'}, \quad j' = 1, \dots, N-1. \quad (A6)$$

The matrix eigenvalue problem is defined by Eq. (A6) and the boundary conditions. For example, for the frequently used Dirichlet boundary conditions,

$$A_0 = A_N = 0, \quad (A7)$$

the sum in the left-hand side of Eq. (A6) is reduced to the range from 1 to  $N-1$ , and a closed set of equations to solve for the eigenvalue  $\lambda$  and eigenvector  $\{A_j, j=1, \dots, N-1\}$  is obtained:

$$\sum_{j=1}^{N-1} [d_{j'j}^{(2)} + \delta_{j'j} v(x_j)] A_j = \lambda A_{j'}, \quad j' = 1, \dots, N-1. \quad (A8)$$

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